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Dirty Bomb Fallout

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DIRTY BOMB FALLOUT

Lawrence Livermore National Laboratory has studied the deposition of caesium on urban concrete after detonation of a mock radiological dispersal device.

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INTRODUCTION

At present, there is a significant need to develop decontamination agents that can be used effectively after detonation of a radiological dispersal device (RDD) in an urban environment. There is also a need for the development of reproducible test surfaces to be used to determine the efficacy of the agent being developed.

Researchers at Lawrence Livermore National Laboratory (LLNL), under the auspices of the US Department of Energy (DoE), conducted a field study to evaluate the deposition of an explosively dispersed radionuclide surrogate (CsCl) on grime-bearing and non-grime-bearing urban surfaces. The goal was to investigate the preparation and contamination of urban surfaces that closely mimic what one would expect to encounter following the detonation of an RDD. Migration of Cs into concrete surfaces was investigated in detail.

Many non-proliferation, security and response organizations that have modeled RDD scenarios use cesium-137, as well as cobalt-60, strontium-90, americium-241 as the most likely RDD agents. Cesium-137 is an isotope of concern for possible use in an RDD due to its potential availability resulting from its widespread legitimate use in construction, geotechnical and medical industrial devices. In some Cs-containing instruments the Cesium-137 is present as the highly dispersible and water soluble salt, cesium chloride (CsCl). In this form Cs is able to rapidly disperse in the environment, as witnessed in the 1987 Goiania accident in Brazil, when inadvertent dispersal of a radiotherapy source resulted in fatalities and injuries.

EXPLOSIVE PREPARATION

Our FY06 field study was conducted at the Lawrence Livermore National Laboratory (LLNL) Site 300 Contained Firing Facility (CFF). For our study, we detonated a mock RDD made using C4 and non-radioactive CsCl. Lab prepared concrete samples (3.8 cm x 7.6 cm cylinders) were made using 4 different conditioning regimes to mimic a range of conditions that may be encountered during an RDD event. This sample set included dry, wet, carbonated and non-carbonated cores with and without the application of urban grime. In addition, intact concrete

samples (13 cm x 13 cm x 5 cm) removed from an urban surface were placed inside the CFF chamber. The samples were placed inside the firing chamber at 3 different distances from the mock RDD device. Following the detonation of the mock RDD, the samples were removed from the firing chamber and selected cores were characterized by laser ablation and scanning electron microscopy.

Two types of concrete samples were placed in the firing chamber; lab prepared 3.8 cm diameter by 7.6 cm length cylindrical cores (Figure 1) and 12.8 cm by 12.8 cm by 5.1 cm thick concrete blocks retrieved intact from an urban transit system subway tunnel. We decided to use lab prepared cores in order to generate a large number of samples with consistent, measurable physical characteristics that could be pre-conditioned in a controlled environment. A total of 300 concrete cores were prepared over a period of 3 months.

Basic Core Preparation

Concrete for the concrete cylinders were prepared in batches that were was sufficient to prepare 25 cores at a time. The concrete ingredients (Portland cement, silica sand and water) were combined in a double planetary mixer and combined for a total of 6 minutes. Cores were poured in two layers in HDPE molds and shaken on an orbital shaker for 20 minutes . Thin welding rods were used to "rod" each layer 20 times to remove any void spaces formed by air bubbles. The cores were manually removed from the molds after 24 hours. Upon removal from the molds, the concrete cores were place in a 19 L bucket of 3 g/L $Ca(OH)_2$ solution to soak for a period of 7 days. After the cores were removed from the $Ca(OH)_2$ solution, they were placed in an empty bucket and thoroughly rinsed with tap water. The cores were labeled by batch and pour number (i.e.: B1-1, B1-2 etc.). After rinsing, 5 cores were removed from each pour, patted dry with a paper towel and weighed. All of the cores were then place in an oven that was pre-heated to 60° C and dried for a period of 48 hours. All 300 basic cores were prepared with this same protocol. Following this initial conditioning period, the cores were further conditioned using one of the following methods: grime addition with accelerated carbonation, grime addition without carbonation, wet conditioning without grime or dry conditioning without grime.

Grime Addition without carbonation

The objective of this conditioning regime was to prepare concrete samples with an attached grime layer similar to that found in urban environments. Two different grime materials were used to condition cores. Thirty-five cores were conditioned with National Institute of Standards and Technology (NIST) Urban Dust (Standard Reference Material (SRM) #1649a)) and 35 cores were conditioned with grime material that was collected from an urban transit system. The transit system grime material was collected from a vent inside of a transit station and was comprised of predominately Fe (40%), S (22%), Ca (15%) and Si (10%). Full characterization of this material is still in progress. The NIST Urban Dust was a well characterized material comprised of inorganic carbon (18%), Fe (3%), S (3%), Pb (1%), Mg (1%), and smaller amounts (<1%) of PAHs, PCBs and pesticides. A 1:1(wt) grime:DI H_2O slurry was prepared in a small glass beaker and 1 mL of slurry was applied to the core surface using a 10 mL disposable plastic syringe. Control cores were prepared by adding 10 mL of DI H_2O to the core surface. The

grimed cores were place in an environmental chamber which held an open container of saturated $Mg(NO_3)_2$ $6H_2O$ solution. The goal was to maintain a relative humidity of ~53% during core conditioning. The cores were maintained under these conditions for a total of 21 days prior to placement in the CFF chamber.

Accelerated Carbonation Conditioning

For this batch of cores, our goal was to prepare concrete cores that would have a measurable carbonation layer to more closely mimic aged concrete that would be found in urban environments. A total of 60 cores (20 NIST, 40 transit grime) were subjected to two applications of grime and two 5 day duration carbonating conditioning periods. Grime slurry was prepared with 0.562 g grime in 22.5 g DI H_2O. Following grime addition, the cores were placed in a vacuum desiccator that contained a saturated KI solution beneath the perforated plate. After the lid was placed on the desiccator, the air in the chamber was evacuated and the headspace was purged with a mixture of 50%N_2/50%CO_2.

Phenolphthalein testing of these cores indicates that no measurable carbonation layer was formed on this set of samples as expected. We believe this is the result of insufficient CO_2 and too little carbonation time. A follow-on series of experiments are planned to develop a more effective accelerated carbonation method.

Wet Conditioning (no grime)

After the 48 hr drying period of the basic cores was completed, a total of 50 cores were subjected to wet conditioning. After the cores were removed from the oven, they were submerged in a container of tap water for a period of 6 days. After the soaking period, the cores removed from the container and placed in a dessicator with a tray of water placed beneath the perforated plate. These cores were held in the dessicator until placement in the sample holder, 2 hours prior to device detonation.

Dry Conditioning (no grime)

After the 48 hr drying period of the basic cores was completed, a total of 50 cores were subjected to dry conditioning without grime addition. After the initial 48 hour drying period had elapsed these cores were dried at 60°C for an additional 72 hours. The cores remained in the drying oven until the day they were placed inside the samples holders, one day before device detonation.

CONFINED FIRING FACILITY SET-UP

The Contained Firing Facility (CFF) at LLNL's Site 300 consists of an approximately 2092 m^3 (73,890 ft^3) firing chamber with dimensions of 17m x 15m x 9m (55' x 50' x 30') that was designed to safely contain detonations of up to 60 kg of high explosives (HE) without release of hazardous materials to the environment. The primary mission of the CFF is to support hydrodynamic testing of both nuclear and conventional weapons systems under the Department of Energy's Stockpile Stewardship program.

Our project team had access to the CFF firing chamber for a total of 6 days. On day one, we set-up the chamber and placed all of our prepared samples with the exception of the wet conditioned samples. On day two the wet conditioned samples were placed in the chamber and the mock RDD was detonated. On days 3, 4, and 5 (weekend) the CFF chamber remained sealed with our sampled inside. On day 6 the CFF chamber was purged and we re-entered the CFF chamber and retrieved our samples.

Sample Placement

In the CFF firing chamber we set three 2m x 2.5m x 5 cm (6'x 8'x 2") steel blast shields at distances of 3, 6, and 9 m (10, 20 and 30 ft) from the mock RDD to serve as immobile surfaces to attach the sample holders to. The sample holders were constructed of high-density aircraft foam that was machined to secure the samples during the detonation over-pressure and facilitate sample removal without disturbing the sample face containing the explosively deposited Cs. The sample holders contained several of each form (wet,dry,grimed etc.) of laboratory prepared samples as well as field collected and analysis specific (e.g. elipsometry) prepared media. The sample holders were attached to the blast shields with metal bands. The vertical sample surfaces were arranged to maximize exposure to the mock RDD. Floor sample holders that contained similar samples placed behind the blast shields and attached to 1m x 1m x 5cm (4'x 4'x 2") steel plates to assess horizontally deposited "fallout" Cs compared to explosively deposited Cs. Sample placement in the chamber is shown below (Figure 2).

Mock RDD

Most CFF experiments are designed with the high explosive (HE) placed over the 150mm thick steel "shot anvil" in the center of the chamber floor. Chamber dimensions in this configuration would not allow us to place samples at 30' from point of detonation. Due to the small amount of HE (1.5 kg C-4) in the mock RDD, we were able to position the mock RDD in a corner of the chamber over a 50mm steel plate. One kg of non-radioactive CsCl was placed in the mock RDD as a surrogate for Cs^{137}. The mock RDD was suspended from an aluminum A-frame at a height of 1 m and aligned with the sample holders attached to the blast shields.

Post-detonation Sample Retrieval

Post-detonation sample retrieval required a modification of CFF chamber re-entry procedures. Typically re-entry requires that a HE handler enter the chamber after 10 volume air changes at 24,000 cfm to verify complete detonation of all HE, to allow for rapid retrieval of diagnostic instrumentation inside the chamber. We were concerned that this high air circulation rate might cause re-entrainment of Cs and disturb the settled "fallout" Cs sample concentrations. We arranged to have the 10 volume air change to proceed at a lower 6000 cfm "operational" rate that the firing chamber is under whenever personnel are present in the chamber. Following the re-entry all-clear procedures, the sample holders were photographed and removed from the firing chamber, and staged in the CFF high bay, covered with 6 mil poly bags, while waiting for release from the facility. Samples identified for thin sectioning and micro-probe characterization had a layer of epoxy and an optical glass plate applied immediately upon sample removal from the

firing chamber. When the samples were approved for release after 30 days, they were transported from Site 300 to LLNL main site for analysis.

RESULTS

Sample characterization at LLNL used several different techniques including laser ablation with inductively coupled plasma spectrophotometry (LA-ICP), scanning electron microscopy (SEM), elipsometry, and ion microprobe.

Preliminary results suggest that significant migration of Cs into concrete surfaces may occur following explosive deposition of Cs on the surface (see data in Figures shown, determined by LA-ICP). The upper, heavy dashed line in Figures 1 and 2 represents the concentration of Cs on the surface of the samples. The double dashed lines at the bottom of both figures represent the Cs concentration in control samples (not place in CFF chamber) plus one standard deviation. The transverse concentrations were measured by splitting the samples vertically and taking a series of laser ablation measurement down the fractured surface.

It appears that Cs migration into the floor sample is evident to a depth of 0.5 cm while in the sample secured to the blast shield, the Cs migration extends at least 1.5 cm below the concrete surface.

It should be noted that the amount of Cs deposited on the concrete surfaces may far exceed what one would expect to encounter following an actual RDD event. We designed a device capable of delivering these higher concentrations to assure that sufficient dispersal of Cs would be achieved to allow the characterization of samples with available analytical instruments.

ACKNOWLEGMENT

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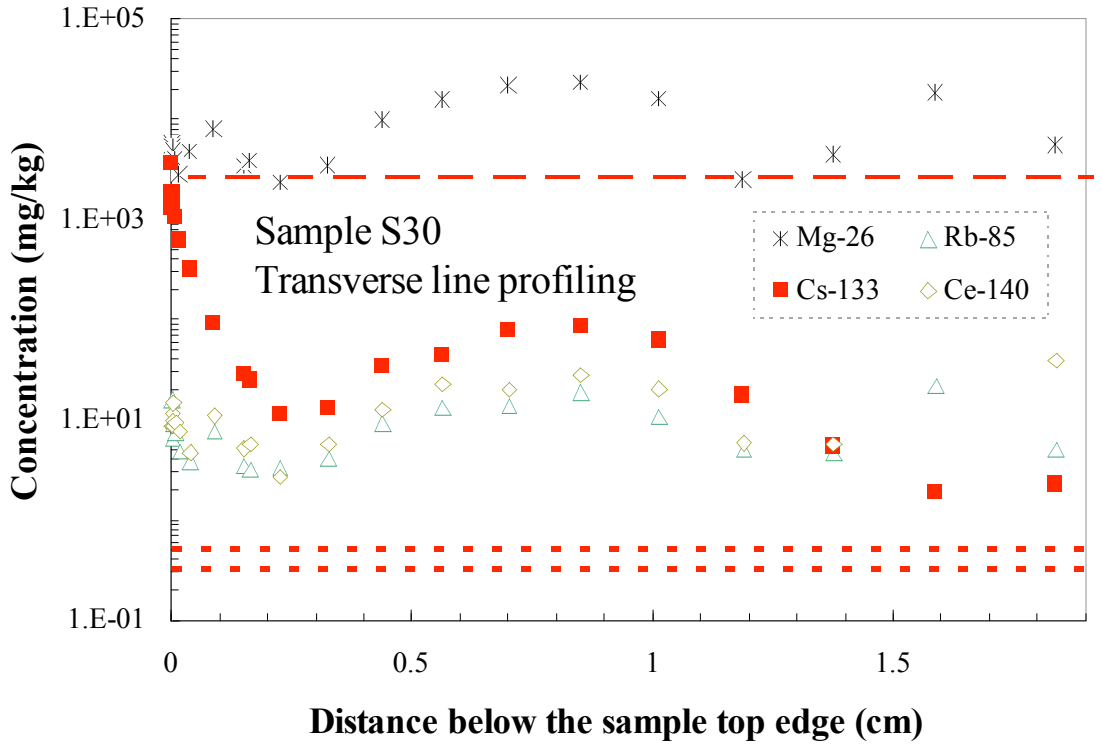
Captions

Figure 1. Cs deposition with depth following detonation of mock RDD. Urban sample placed on blast shield located 9 m from mock RDD.

Figure 2. Cs deposition with depth following detonation of mock RDD. Urban concrete sample placed on floor 9 m from mock RDD.

Figure 1


1.E+05
1.E+03
1.E+01
1.E-01
Concentration (mg/kg)
Sample S30
Transverse line profiling
Mg-26
Rb-85
Cs-133
Ce-140
0
0.5
1
1.5
Distance below the sample top edge (cm)

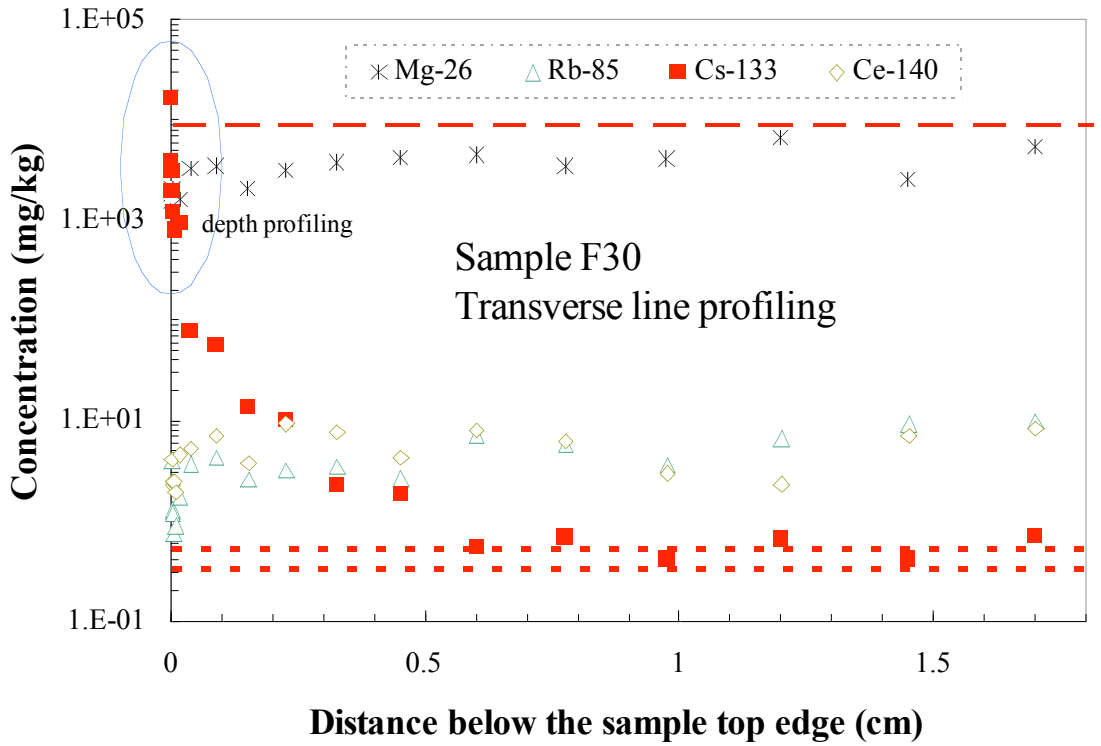

1.E+05
1.E+03
1.E+01
1.E-01
Concentration (mg/kg)
Mg-26
Rb-85
Cs-133
Ce-140
depth profiling
Sample F30
Transverse line profiling
0
0.5
1
1.5
Distance below the sample top edge (cm)

Figure 2